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(54) Title: SYNERGISTIC HERBICIDAL COMPOSITION AND METHOD OF USE THEREOF

(57) Abstract

A synergistic herbicidal composition comprising (a) 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione and (b) 2-chloro-4-ethylamino-6-isopropylamino-S-triazine. Also disclosed is a method of controlling undesirable vegetation by applying an effective amount of such composition to the locus of the vegetation to be controlled.

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SYNERGISTIC HERBICIDAL COMPOSITION AND METHOD OF USE THEREOF

Field of the Invention

In one aspect the present invention is directed to a synergistic herbicidal composition comprising (a) 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione; and (b) 2-chloro-4-ethylamino-6-isopropylamino-S-triazine. In another aspect, the present invention is directed to a method of controlling the growth of undesirable vegetation by applying an herbicidally effective amount of such synergistic composition to the locus of such vegetation.

Background of the Invention

The protection of crops from weeds and other vegetation which inhibit crop growth is a constantly recurring problem in agriculture. To help combat this problem, researchers in the field of synthetic chemistry have produced an extensive variety of chemicals and chemical formulations effective in the control of such unwanted growth. Chemical herbicides of many types have been disclosed in the literature and a large number are in commercial use.

In some cases, active herbicides have been shown to be more effective in combination than when applied individually. The result is often termed "synergism", since the combination demonstrates a potency or activity level exceeding that which it would be expected to have, based on a knowledge of the individual potencies of the components. The present invention resides in the discovery that certain cyclohexanediones and 2-chloro-4-ethylamino-6-isopropylamino-S-triazine-(atrazine), already known individually for their herbicidal

potency, display a synergistic effect when applied in combination.

The compounds forming the combination which is the subject of the present invention are independently known in the art for their effects on plant growth. Thus, 2-chloro-4-ethylamino-6'-isopropylamino-S-triazine, commonly known as atrazine, is commercially sold under various trade names, and is described in the Herbicide Handbook of the Weed Science Society of America, 5th Edition, 1983; 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione is disclosed in United States Patent 5,006,158 to Carter et al; and 2-(2'-nitro-4'-methylsulfonyloxybenzoyl-1,3-cyclohexanedione is disclosed in United States Patent 5,089,046 to Lee et al.

Description of the Invention

In one aspect, the present invention is directed to a synergistic herbicidal composition comprising (a) 2-(2'-nitro-4'methylsulfonylbenzoyl)-1,3-cyclohexanedione ("NMSC") or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione ("NMSOC"); and (b) 2-chloro-4-ethylamino-6-isopropylamino-S-triazine.

In another aspect, this invention is directed to a method of controlling undesirable vegetation applying to the locus of such vegetation a synergistic composition comprising (a) 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione; and (b) 2-chloro-4-ethylamino-6-isopropylamino-S-triazine.

The terms "synergism" and synergistic" are used herein to convey the result observed when a combination of herbicides demonstrates a potency in excess of that which the combination would be expected to produce based upon the potencies of each herbicide applied separately.

The term "herbicide" is used herein to denote a compound which controls or modifies the growth of plants. The term "herbicidally effective amount" is used to indicate the quantity of such a compound or combination of such compounds which is capable of producing a controlling or modifying effect. Controlling or modifying effects include all deviations from natural development, for example: killing, retardation, leaf burn, albinism, dwarfing and the like. The term "plants" refers to all physical parts of a plant, including seeds, seedlings, saplings, roots, tubers, stems, stalks, foliage and fruits.

In the compositions of this invention, the weight ratio of component (a) to component (b) at which the herbicidal response is synergistic, lies within the range of between about 1:50 and about 1:1, and is preferably between about 1:40 and about 4:5.

Application rates will depend upon the particular plant species and degree of control desired. In general, the compositions of the invention are most efficiently employed at a rate of 0.001 to 5 pounds per acre (0.001 to 5 kilograms per hectare).

The compositions of this invention are useful as herbicides, demonstrating synergistic activity for the control of undesirable vegetation. The compositions can be formulated in the same manner in which herbicides are generally formulated. The compounds may be applied either separately or combined as part of a two-part herbicidal system.

The object of the formulation is to apply the compositions to the locus where control is desired by a convenient method. The "locus" is intended to include soil, seeds, and seedlings, as well as established vegetation.

The composition employed in the practice of the present invention can be applied in a variety of ways known to

those skilled in the art, at various concentrations. The composition is useful in controlling the growth of undesirable vegetation by pre-emergence or post-emergence application to the locus where control is desired. In practice, the composition is applied as a formulation containing the various adjuvants and carriers known to or used in the industry for facilitating dispersion. The choice of formulation and mode of application for any given compound may affect its activity, and selection will be made accordingly. The compositions of the invention may thus be formulated as granules, as wettable powders, as emulsifiable concentrates, as powders or dusts, as flowables, as solutions, suspensions or emulsions, or in controlled-release forms such as microcapsules. These formulations may contain as little as about 0.5% to as much as amount 95% or more by weight of active ingredient. The optimum amount for any given compound will depend upon the nature of the seeds or plants to be controlled.

Wettable powders are in the form of finely divided particles which disperse readily in water or other liquid carriers. The particles contain the active ingredient retained in a solid matrix. Typical solid matrices include fuller's earth, kaolin clays, silicas and other readily wet organic or inorganic solids. Wettable powders normally contain about 5% to about 95% of the active ingredient plus a small amount of wetting, dispersing, or emulsifying agent.

Emulsifiable concentrates are homogeneous liquid compositions dispersible in water or other liquid, and may consist entirely of the active compound with a liquid or solid emulsifying agent, or may also contain a liquid carrier, such as xylene, heavy aromatic naphthas, isophorone and other non-volatile organic solvents. In use, these concentrates are dispersed in water or other liquid and normally applied as a spray to the area to be treated. The amount of active ingredient may range from about 0.5% to about 95% of the concentrate.

Granular formulations include both extrudates and relatively coarse particles, and are usually applied without dilution to the area in which suppression of vegetation is desired. Typical carriers for granular formulations include sand, fuller's earth, attapulgite clay, bentonite clays, montmorillonite clay, vermiculite, perlite and other organic or inorganic materials which absorb or which can be coated with the active compound. Granular formulations normally contain about 5% to about 25% active ingredients which may include surface-active agents such as heavy aromatic naphthas, kerosene and other petroleum fractions, or vegetable oils; and/or stickers such as dextrins, glue or synthetic resins.

Dusts are free-flowing admixtures of the active ingredient with finely divided solids such as talc, clays, flours and other organic and inorganic solids which act as dispersants and carriers.

Microcapsules are typically droplets or granules of the active material enclosed in an inert porous shell which allows escape of the enclosed material to the surroundings at controlled rates. Encapsulated droplet are typically about 1 to 50 microns in diameter. The enclosed liquid typically constitutes about 50 to 95% of the weight of the capsule, and may include solvent in addition to the active compound. Encapsulated granules are generally porous granules with porous membranes sealing the granule pore openings, retaining the active species in liquid form inside the granule pores. Granules typically range from 1 millimeter to 1 centimeter, preferably 1 to 2 millimeters in diameter. Granules are formed by extrusion, agglomeration or prilling, or are naturally occurring. Examples of such materials are vermiculite, sintered clay, kaolin, attapulgite clay, sawdust and granular carbon. Shell or membrane materials include natural and synthetic rubbers, cellulosic materials, styrene-butadiene copolymers, polyacrylonitriles, polyacrylates, polyesters, polyamides, polyureas, polyurethanes and starch xanthates.

Other useful formulations for herbicidal applications include simple solutions of the active ingredient in a solvent in which it is completely soluble at the desired concentration, such as acetone, alkylated naphthalenes, xylene and other organic solvents. Pressurized sprayers, wherein the active ingredient is dispersed in finely-divided form as a result of vaporization of a low boiling dispersant solvent carrier may also be used.

Many of these formulations include wetting, dispersing or emulsifying agents. Examples are alkyl and alkylaryl sulfonates and sulfates and their salts; polyhydric alcohols; polyethoxylated alcohols; esters and fatty amines. These agents when used normally comprise from 0.1% to 15% by weight of the formulation.

Each of the above formulations can be prepared as a package containing the herbicide together with other ingredients of the formulation (diluents, emulsifiers, surfactants etc.). The formulations can also be prepared by a tank mix method, in which the ingredients are obtained separately and combined at the grower site.

These formulations can be applied to the areas where control is desired by conventional methods. Dust and liquid compositions, for example, can be applied by the use of power-dusters, boom and hand sprayers and spray dusters. The formulations can also be applied from airplanes as a dust or a spray or by rope wick applications. To modify or control growth of germinating seeds or emerging seedlings, dust and liquid formulations can be distributed in the soil to a depth of at least one-half inch below the soil surface or applied to the soil surface only, by spraying or sprinkling. The formulations can also be applied by addition to irrigation water. This permits penetration of the formulations into the soil together with the irrigation water. Dust compositions, granular compositions or liquid formulations applied to the surface of the soil can be distributed below the surface of the soil

by conventional means such as discing, dragging or mixing operations.

EXAMPLES

The following Examples are intended to further illustrate the present invention and are not intended to limit the scope of this invention in any manner whatsoever.

In all three of the following examples, test plots were compared to untreated plots and rated visually in terms of percent control ranging from 0% to 100%, with 0% representing no injury to the plant species and 100% representing complete kill of all plants. All types of plant injury were taken into consideration.

The observed results from the herbicide treatments on each species for Examples I, II and III are shown in TABLES I, II and III, respectively. The observed ratings for the individual herbicide treatments are listed along with the expected ("E") and observed ("O") results from the mixtures of NMSC and atrazine. The expected results were derived from the control data using Limpel's formula (Limpel et al., 1962, "Weed Control by Dimethylchloroterephthalate Alone and in Certain Combinations", Proc. NEWCC., Vol. 16:48-53:

$$E = X + Y - \frac{XY}{100}$$

where X = observed percent injury when one of the herbicides is used alone, and

Y = observed percent injury when the other herbicide is used alone.

EXAMPLE I

In Holambra, Brazil, plots were planted with the following weed species: *Echinochloa crus-galli* ("ECHCG"); *Brachiaria plantaginea* ("BRAPL"); *Digitaria ciliaris* ("DIGAD"); *Cenchrus echinatus* ("CCHEC"); *Euphorbia heterophylla* ("EPHHL"); *Bidens pilosa* ("BIDPI"); *Sida rhumbifolia* ("SIDRH") and *Amaranthus retroflexus* ("AMARE"). When such weeds reached the 3-5 leaf stage, they were treated with NMSC; atrazine; or NMSC + atrazine, at the rates indicated in Table I. Injury to the species in each plot was rated twenty-eight days after treatment. The results of such testing, as the mean of two replications, are summarized in TABLE I.

TABLE I

NMSC*	ECHCG	BRAPL	DIGAD	CCHEC	EPHHL	BIDPI	SIDRH	AMARE	IPUAO
18	0	0	3	0	42	53	47	33	23
35	23	0	0	5	43	57	72	45	35
70	30	8	20	0	47	69	82	50	53
140	45	13	52	3	60	91	99	72	67
Atrazine*									
250	0	0	0	0	30	30	10	23	18
500	0	0	0	13	47	45	30	40	33

	ECHCG		BRAPL		DIGAD		CCHEC		EPHHL		BIDPI		SIDRH		AMARE		IPUAO	
NMSC + Atr*	E	O	E	O	E	O	E	O	E	O	E	O	E	O	E	O	E	O
18 + 250	0	13	0	0	3	23	0	0	59	62	67	80	52	88	48	91	37	64
35 + 250	23	33	0	13	0	20	5	8	60	58	70	91	75	98	58	89	47	68
70 + 250	30	45	8	12	20	33	0	12	63	67	78	100	84	100	61	93	61	79
140 + 250	45	91	13	32	52	72	3	13	72	80	94	100	99	100	88	100	73	90
18 + 500	0	27	0	3	3	13	13	12	69	53	74	90	63	97	60	92	48	65
35 + 500	23	57	0	13	0	18	17	7	70	71	76	100	80	99	67	95	55	82
70 + 500	30	67	8	12	20	40	13	7	72	69	83	100	87	100	70	100	69	95
140 + 500	45	92	13	40	52	63	16	13	79	90	95	100	99	100	83	97	78	98

*Application rates in grams per hectare.

EXAMPLE II

Near Vienna, Austria, plots were planted with seeds of the following species: *Amaranthus retroflexus* ("AMARE"); *Chenopoduim album* ("CHEAL"); *Chenopoduim hybruim* ("CHEAL") and *Datura stramonum* ("DATST"). When such plants reached the 3-5 leaf stage, they were treated with NMSC alone; atrazine alone; or NMSC + atrazine, at the rates indicated in Table II.

Thirty-five days after such application, the treated plants were compared to untreated plants. The results of such testing, as the mean of 3 replications, along with the results expected using the Limpel Formula, are summarized below in Table II.

TABLE II

NMSC Alone*	AMARE	AMACH
12.5	52	47
25	65	63
50	80	80
100	87	83
200	98	97
Atrazine Alone*		
250	20	18
500	58	53

NMSC + Atrazine*	AMARE E	AMARE O	AMACH E	AMACH O
12.5 + 250	62	85	57	85
25 + 250	72	96	70	95
50 + 250	84	99	84	99
100 + 250	90	99	86	98
200 + 250	98	100	98	100
12.5 + 500	80	98	75	98
25 + 500	85	99	83	98
50 + 500	92	98	91	98
100 + 500	95	99	92	99
200 + 500	99	100	99	100

*Application rates in grams per hectare.

Due to essentially complete control of CHEAL, CHEHY and DATST by NMSC and/or atrazine alone, little or no evidence of synergy was observed with respect to these species.

EXAMPLE III

In plots located in Illinois, Iowa, Indiana, Kansas, Minnesota, South Dakota and Missouri (three replications each), seeds of the following weed species occurred: *Xanthuim strumarium* ("XANST"); *Setaria faberi* ("SETFA"); *Abutilon theophrasti* ("ABUTH"); *Chenopodium album* ("CHEAL"); *Polygonum pensylvanicum* ("POLPY"), *Solanium ptycanthum* ("SOLPT"); *Amaranthus retroflexus* ("AMARE"); and/or *Polygonum persicaria* ("POLPE"). The plots were treated with NMSC alone; atrazine alone; or NMSC + atrazine, at the rates indicated in Table III.

The results of each treatment (as a mean of the replications and location) along with the expected results from the Limpel formula are presented below in Table III.

Table III

NMSC rate lb/a	atrazine rate lb/a	XANST % control observed and expected, by location								SETFA	
		KS		MN		IA		IL		IL	
0.063	--	86		88		52		45		10	
0.125	--	89		99		85		75		10	
0.179	--	--		--		92		--		--	
0.25	--	93		99		97		100		42	
--	0.5	90		25		20		42		61	
--	1	90		56		68		50		53	
		E	Q	E	Q	E	Q	E	Q	E	Q
0.063	0.5	99	95	91	99	62	78	68	93	65	88
0.125	0.5	99	96	99	99	88	90	86	94	65	72
0.179	0.5	--	--	--	--	94	95	--	--		--
0.25	0.5	--	--	--	--	--	--	--	--		--
0.063	1	--	--	--	--	--	--	50	88		--
0.125	1	--	--	--	--	--	--	88	100		--

no. of leaves:	4-5	1-4	2-6	4+
days after treatment that test was rated	36	42	55	14

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Little or no synergy of ABUTH, CHEAL, POLPY, SO2PT, AMARE or POLPE was observed due to the almost complete control exhibited by NMSC alone at the rates tested. With respect to AMARE, it is noted that this response (relative to the testing summarized in TABLE II) is not contradictory given the difference in hardiness of the European and American Species.

What is claimed is:

1. A synergistic herbicidal composition comprising (a) 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione; and (b) 2-chloro-4-ethylamino-6-isopropylamino-S-triazine.

2. A composition in accordance with claim 1 wherein the weight ratio of component (a) to component (b) is between about 1:50 and about 1:1.

3. A composition in accordance with claim 1 wherein the weight ratio of component (a) to component (b) is between about 1:40 and about 4:5.

4. A composition in accordance with claim 1 wherein component (a) is 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione.

5. A method of controlling undesirable vegetation comprising applying to the locus of such vegetation an effective amount of a synergistic herbicidal composition comprising (a) 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione or 2-(2'-nitro-4'-methylsulfonyloxybenzoyl)-1,3-cyclohexanedione; and (b) 2-chloro-4-ethylamino-6-isopropylamino-5-triazine.

6. A method in accordance with claim 6 wherein the weight ratio of component (a) to component (b) is between about 1:50 and about 1:1.

7. A method in accordance with claim 6 wherein the weight ratio of component (a) to component (b) is between about 1:40 and about 4:5.

8. A method in accordance with claim 5 wherein component (a) is 2-(2'-nitro-4'-methylsulfonylbenzoyl)-1,3-cyclohexanedione.

9. A method in accordance with claim 5 wherein said composition is applied postemergence.

INTERNATIONAL SEARCH REPORT

Int onal Application No
PCT/GB 95/00721

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 A01N43/70 //(A01N43/70,41:10,41:04)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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Y	EP,A,0 354 047 (MAY & BAKER LTD) 7 February 1990 see page 2, line 3 - line 12 see column 2, line 17 - line 20 see page 2, line 33 - line 40 ---	1-9
Y	EP,A,0 230 596 (STAUFFER CHEMICAL CO) 5 August 1987 see page 2, line 1 - line 12 ---	1-9

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
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Name and mailing address of the ISA	Authorized officer
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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PCT/GB 95/00721

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		DE-A-	3688844	09-09-93
		DE-T-	3688844	05-01-94
		DE-D-	3688982	07-10-93
		DE-T-	3688982	28-04-94
		EP-A,B	0347950	27-12-89
		EP-A,B	0350079	10-01-90
		ES-T-	2058416	01-11-94
		ES-T-	2058417	01-11-94
		US-A-	4759794	26-07-88
EP-A-0336898	11-10-89	AU-A-	3238289	05-10-89
		JP-A-	2006426	10-01-90
		SU-A-	1760982	07-09-92
		US-A-	5089046	18-02-92
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		US-A-	5416061	16-05-95
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